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RADIONUCLIDE ANALYSIS OF LARGE NUMBERS OF FOOD AND WATER SAMPLES

Ester Ferri, et al

Public Health Service
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Radiological Health

RADIONUCLIDE ANALYSIS OF LARGE NUMBERS OF FOOD AND WATER SAMPLES

U. S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE

Public Health Service

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RADIONUCLIDE ANALYSIS OF LARGE NUMBERS OF FOOD AND WATER SAMPLES

by

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Public Health Service

DIVISION OF RADIOLOGICAL HEALTH
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PREFACE

As a part of their responsibilities for gathering and assessing trends in environmental radiation levels, the laboratories of the Division of Radiological Health process a large number and variety of samples collected under the various nationwide networks of the Public Health Service. In July 1964, the staff of the Northeastern Radiological Health Laboratory published a two-part report,[1] recording experiences in developing and placing that facility in operation and outlining some of the major considerations in the analysis of large numbers of fluid milk samples. The present report is an extension of this series and relates procedures found satisfactory for large scale processing of food and water samples for radionuclide measurements.

The first section of this report covers the processing of individual food items and total diets; the second covers the handling of drinking and surface water samples. Included in each section are the necessary requirements for equipment and personnel—all expressed in terms of sample workload. Through this approach, the values can be scaled upward or downward to fit a given requirement. Although individual situations will require modifications of these approaches, it is hoped that the data presented will be of help to individuals and groups faced with responsibilities for the development of similar programs within State and local health departments.

The use in this report of trade names of commercially available products is only to indicate that the particular product is being used in the laboratory. Such use of the name or the product does not constitute an endorsement of the product by the Public Health Service or the U.S. Department of Health, Education, and Welfare, and does not imply that there are no other suitable products available commercially.

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[1] An Environmental Surveillance Laboratory for Radionuclide Analysis, Environmental Health Series, Public Health Service Publication No. 999–RH–7, U.S. Government Printing Office (July 1964).

ABSTRACT

As part of the responsibilities of the Division of Radiological Health for environmental surveillance, large numbers of samples are analyzed for gross and specific radioactivity. The processing of food and water samples, as conducted at the Northeastern Radiological Health Laboratory, is described in this report. Included are manpower and equipment requirements, and technical procedures for measurements of appropriate radioactivities. Procedures described include techniques for a series of radionuclides, as well as certain stable elements in food, and for gross alpha and beta determinations, gamma spectral analyses, and radium 226 and strontium 90 in water.

SECTION I

RADIONUCLIDE ANALYSIS OF LARGE NUMBERS OF FOOD SAMPLES

Esther Ferri

INTRODUCTION

To estimate the intake of radioactive materials by humans, the Division of Radiological Health conducts a comprehensive program for the determination of specific radionuclides in selected food items and total diets. The testing of individual foods is designed to provide data on the major dietary contributors, on variations in their nuclide content from one geographical location to another, and on factors influencing these variations. Data from samples of the total diet are used to estimate the total radionuclide intakes of selected population groups and to determine the variations in different geographical locations.

The present levels of radioactive materials in most foods have been extremely low, requiring the processing of large samples to obtain the desired data. In addition to the determination of radionuclides of health significance, supplementary data on stable elements having similar chemical properties are necessary because these affect the behavior of radionuclides in the body. The program in operation at the Northeastern Radiological Health Laboratory to obtain data for the food surveillance systems of the Division of Radiological Health is described in this report. This example may also assist others in planning similar programs. For detailed information on the establishment of the facility in which the described work was performed, reference is made to a report by Baratta, Parks, and Moeller (*1*).

ORGANIZATIONAL ASPECTS

SPECIFIC FOOD ITEMS

Specific food items are collected at major marketing centers throughout the United States and shipped to the Northeastern Radiological Health Laboratory. These samples are collected quarterly and are representative of food products from various sections of the country. Information on the distribution of food items in the major marketing centers is obtained from the Department of Agriculture. The types of samples to be assayed were determined from a "market basket" consumption survey obtained from the Bureau of the Census.

Specific food items to be analyzed may be categorized as follows: Foods that grow beneath the ground surface, such as potatoes and carrots, whose source of radioactivity is from fallout via translocation and uptake from the soil; those that grow at the surface, such as cabbage and lettuce, which may contain activity directly from fallout as well as from the soil; those that grow above ground level, such as coffee, apples, and oranges, which include contamination from fallout and soil uptake; food products of animals origin, such as eggs, milk, and meat, in which nuclides are present subsequent to ingestion and metabolism; and processed products, such as oleomargarine, instant coffee, and bread, which may become contaminated by additives in the course of preparation.

TOTAL DIET SAMPLES

Total diet samples are obtained from institutions housing children between the ages of 10 and 17 years. A Public Health Service representative and representatives of appropriate State and local health agencies request each participating institution to collect a sample diet for 1 week (21 individual meals and snacks) during each month (*2*).

Detailed instructions for collecting and storing samples, and for the shipment of containers, are furnished by the analytical laboratory. A description and the purpose of each item furnished the institutions are presented in table 1.

Table 1. Monthly supply of containers for collection of diet samples

Item	Quantity	Purpose
Polyethylene bottle, 1 gal	1	Collection of drinking water
Shipping case, padded	1	Shipment of water
Plastic jars, 1 gal	3	1 for milk; 1 (2 if needed) for other liquids (broth, juices, etc.)
Plastic bag	30	Collection of individual meals plus snacks
Cardboard cartons, 1 gal	7	Meal and snack collection each day
Shipping carton, corrugated, 15- by 15- by 26 in	1	Shipment of total milk and food

A form is included for recording the foods and beverages served at all meals and snacks. During the period of collection, the food is preserved by refrigeration. For shipment, the diet samples are

packed in dry ice and sent via air express to the laboratory. The diet sheet is included in the package and the notification of shipment is mailed separately at the same time. The usual weight of foods including milk received ranges from 9 to 12 kilograms. Although individual records from the participating institutions are kept confidential, summaries and compilations are available.

FLOW OF SAMPLES FOR ANALYSIS

All samples received in the laboratory, except drinking water, require preparation prior to analysis. Figure 1 shows the progress of samples through the laboratory from receipt to final reporting. Samples arriving at the receiving room are given an identifying number, and a record is made of the date and place of collection. The samples are

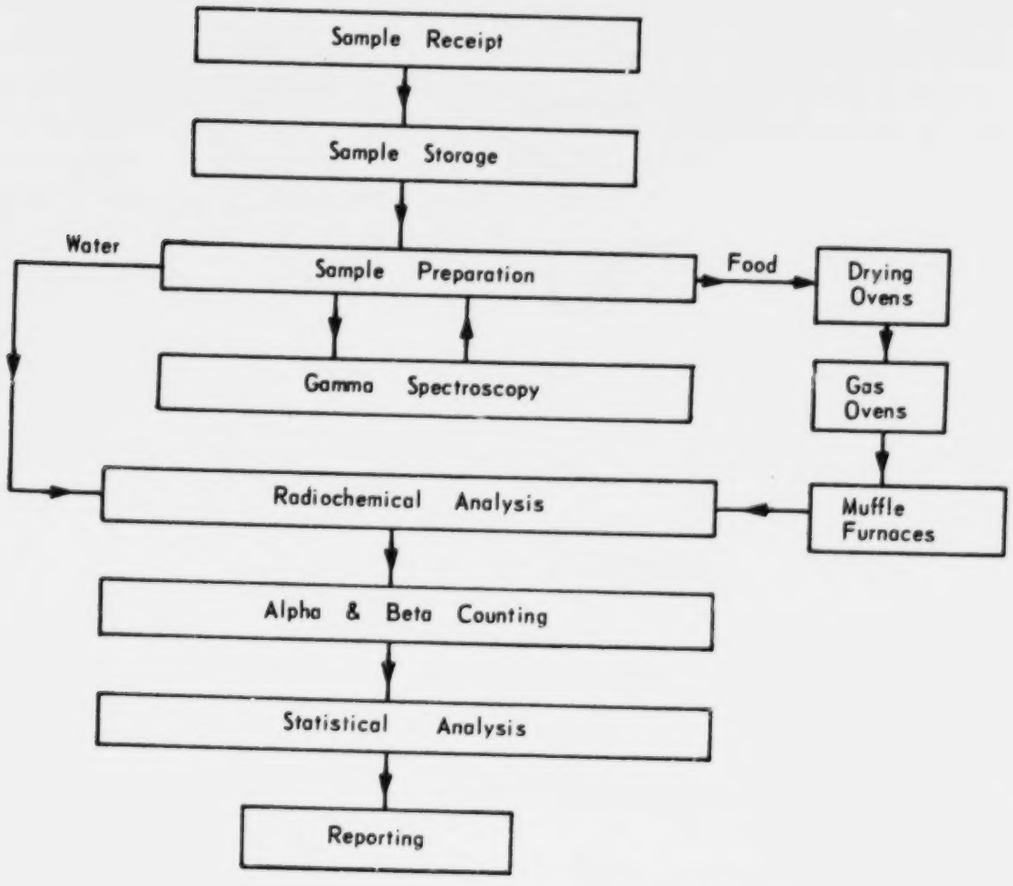

Figure 1. Flowchart of sample route

then sent to the sample-preparation room. Specific food samples are prepared by separating waste materials (the portions not normally ingested, such as skins, shells, and cores) from the edible portions. Total diet samples should contain only edible materials. The edible and waste portions are homogenized separately, and a representative aliquot weighed and submitted for gamma analysis. Liquids other than water are included in the edible portion. After gamma analysis, the samples are returned to the sample-preparation room, dried to determine moisture content, and then moved through ovens and furnaces at increasingly higher temperatures until a gray or white ash is obtained. It has been found that ash represents about one-hundredth of the weight of the original sample and contains all the nuclides of interest to be analyzed chemically. The ash is submitted to the general chemistry laboratory for stable chemical and radiochemical analyses. Any remaining ash is returned to the sample-preparation room and stored for 6 months to provide a sample source for use should the original be lost. The same portion of the original sample is used throughout the entire process, since this avoids confusion as to possible duplication of numbers and gives final results on the same aliquot. Any remaining portions of fresh raw material are retained as additional sample sources until final results are complete.

TECHNICAL ASPECTS

METHODOLOGY

The radionuclides of interest in foods are cesium 137, cerium 141–144, zinc 65, zirconium-niobium 95, barium 140, ruthenium 103–106, iodine 131, strontium 89–90, yttrium 90, potassium 40, and total radium. Stable nuclides of interest are calcium, phosphorus, potassium, and strontium.

The radionuclides of cesium, cerium, zinc, zirconium, barium, ruthenium, potassium, and iodine are determined by gamma spectrometry in a multichannel analyzer equipped with a 4- by 4-inch NaI(Tl) crystal in a shielded chamber. By using matrix equations [similar to those developed by Hagee, *et al.* (*3*) for milk, and described in a PHS training manual (*4*)], the concentration of nuclides is measured. Stable potassium is calculated from the potassium 40 content.

The radionuclides separated and analyzed by radiochemical techniques are radium, strontium 89–90, and yttrium 90. Total radium concentration is determined in an internal gas-flow proportional counter. Strontium 89–90 and yttrium 90 (all pure beta emitters) are determined by counting in a low-background beta counter.

The ash, obtained from the raw material which has been gamma scanned, is solubilized by acid leach and alkali fusion and adjusted to a known volume. From this, aliquots are taken for the remaining analyses. Calcium is determined by oxalate precipitation-permanganate titration (*5*). Phosphorus is determined colorimetrically as the yellow molybdovanadophosphoric acid by comparison to known standards using a model DU spectrophotometer (*6*). Although it is not done routinely at this laboratory, stable strontium may be determined by neutron activation. Strontium 89, strontium 90, and total radium are determined on the same aliquot. These radionuclides are concentrated by carbonate precipitation using strontium and barium carriers, and then purified by rare-earth scavenging and nitrate separations.

The barium (radium) is separated from strontium by chromate precipitation and counted for alpha activity in a gas-flow proportional counter. The portion containing the strontium nuclides is set aside for a period of at least 5 days to allow ingrowth of yttrium 90, the daughter product of strontium 90. At the end of the ingrowth period, the yttrium 90 is separated by solvent extraction, precipitated as the oxalate, and counted for beta activity in a low-background counter. Total radiostrontium is determined by carbonate precipitation and beta counting; strontium 89 is obtained by difference (*7*, *8*) (see fig. 2). Radiochemical purity is determined by ingrowth and decay curves, and flame photometry is employed to determine chemical purity of precipitates.

Drinking-water samples received as part of the total diets are analyzed using the above procedures but do not require any preparation prior to analysis. They are gamma scanned and then treated as solubilized ash solutions for radiochemical analysis.

QUALITY CONTROL

To insure reliability of techniques and results, an intralaboratory quality control program is maintained. Twenty percent of the samples are resubmitted, with new identification, as blind checks. These results are compared with the originals, and the overall evaluation of the precision of the techniques is established. To establish accuracy of the results, samples spiked with known amounts of nuclides are submitted periodically.

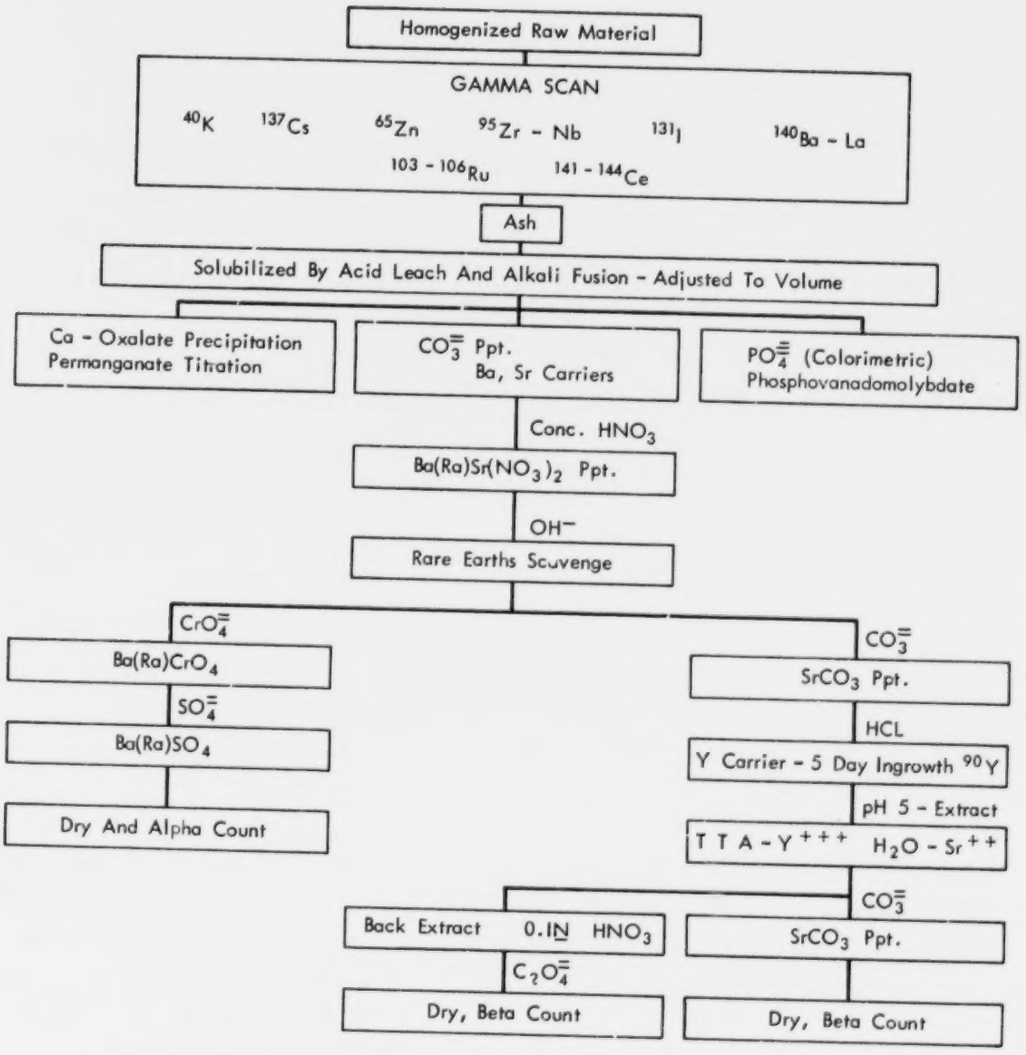

Figure 2. Flowchart—Radiochemical and chemical techniques

OPERATIONAL ASPECTS

SAMPLE PREPARATION

Equipment employed (figs. 3A through 3E) for storage and preparation of food samples is listed in table 2. Operating conditions, sample capacities, and quantities are included for each item.

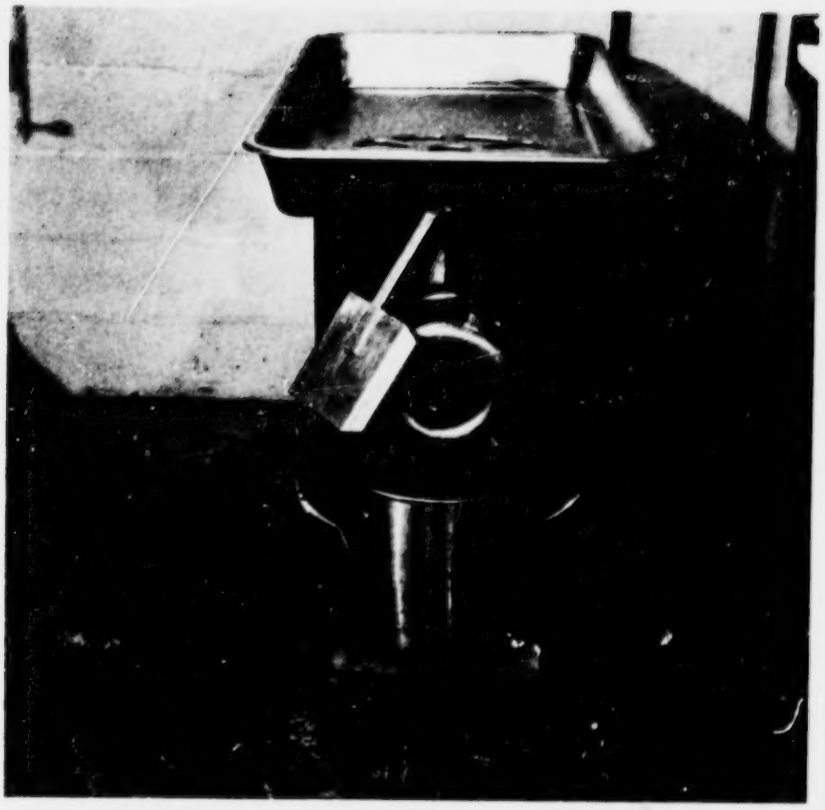

Figure 3A. Meat grinder

Figure 3B. Walk-in refrigerator freezer (interior)

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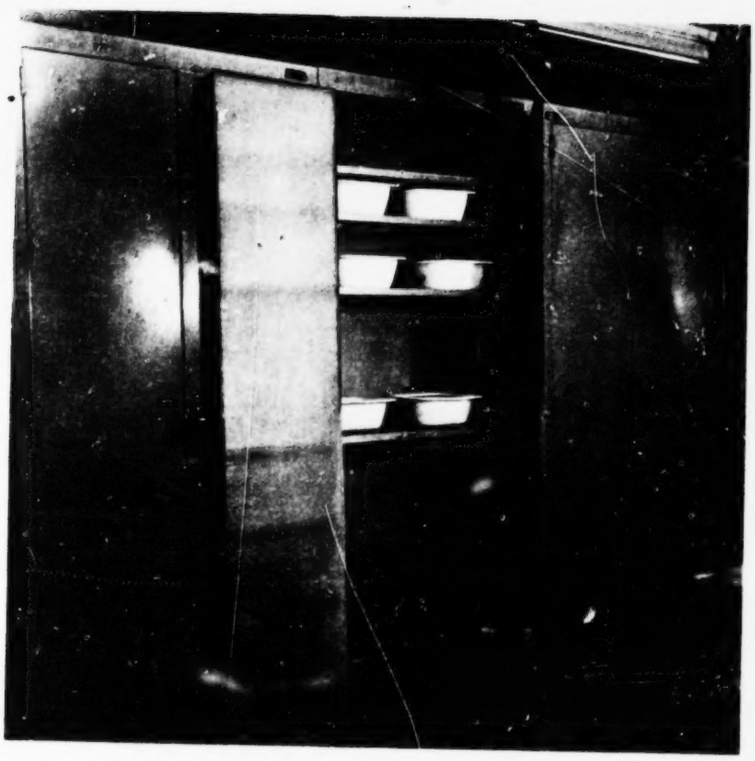

Figure 3C. Drying ovens

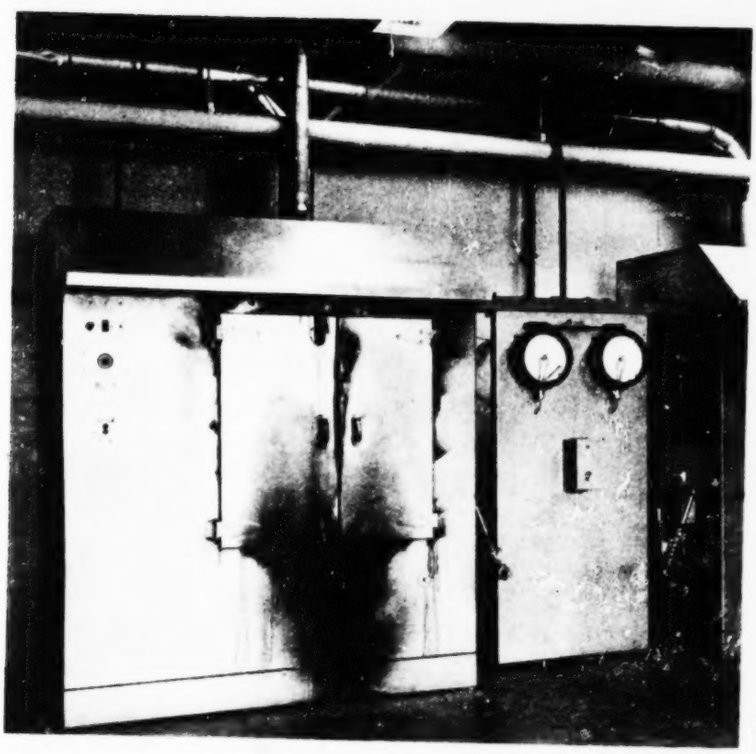

Figure 3D. Gas furnaces

Figure 3E. Muffle furnaces

Time Requirements

The man-hours and elapsed time required for sample preparation are presented in table 3. Time studies were conducted for two samples to simulate actual operating conditions for running an original sample plus a duplicate to maintain quality control. Preparation of a single sample would not halve the time requirements.

The elapsed time required to complete sample preparation is 1 week. Based on the man-hours available in 1 week (40), and two samples requiring 2¾ hours, one person could prepare 30 samples per week. Capacities of drying ovens and furnaces operating simultaneously total 1,056 pounds. The capacity of the ovens and furnaces is sufficient to dry and ash the samples prepared continuously by four people.

SAMPLE ANALYSIS

Gamma Analysis

The laboratory uses two 400-channel analyzers split into four 200-channel units for gamma scanning of food. Samples are counted for 50 minutes. Limits of detectability for the nuclides of interest are given in table 4.

The counting time capacity for the four analyzers is 130 hours per 40-hour week or 130 samples per 40-hour week with 2 hours per day allowed for instrument repair, calibration, background counting, sample changing, and related procedures.

Table 2. Sample preparation equipment at NERHL

Item	Quantity	Operating conditions	Total sample capacity
Freezer-refrigerator walk-in	1	Refrigerator, 35° F Freezer, 14° F	49,000 lb
Upright freezer	3	Temperature, 14° F	2,100 lb
Refrigerator [a]	2	Temperature, 35° F	840 lb
Meat chopper [a]	1	Varying blade attachment	1,000 lb/day
Waring blender [a]	3	—	1,008–2,008 lb/day
Drying ovens [a]	3	Temperature range, 105°–110° C	720 lb
Electric furnace	2	Maximum temperature, 325° C	64 lb
Gas furnace	2	Programed for maximum temperature up to 950° F (510° C)	144 lb
Muffle furnace	4	Usual operating temperature, 1,112° F (600° C)	128 to 1,056 lb
Scales	2	—	0–25 kg, 0–75 lb
Balance	1	—	0–1,610 g
Ash pans	Varies	Pyrex-drying and low temperature ashing	3,500 g
		Pyro-Ceram High-temperature ashing	200–300 g
		Coors-evaporator	

[a] Optional equipment, not essential but serves as additional aids in processing large numbers of samples.

Table 3. Time required for sample preparation [a]

Procedure	Man-hours	Actual elapsed time (hours)
Preparation: Defrosting, weighing, homogenizing	½	48
Drying of sample	¾	48
Ashing:		
Low temperature	½	12
High temperature	½	48
Weighing	½	
	2¾	156
		2¾
		Total: 158¾

[a] Data are given for two samples.

Table 4. Minimum detectable limits of various nuclides using a gamma spectroscopy system [a]

Nuclide	pCi/kg
Cesium 137	12
Cerium 144	58
Zinc 65	15
Zirconium 95	3
Barium 140	8
Ruthenium 103–106	50
Iodine 131	9
Potassium 40	80 (0.06 g/kg equivalent)

[a] 3.5-kilogram samples in Marinelli beakers.

Radiochemistry

Preparation for counting

Exclusive of counting and calculation time, stable nuclide determinations and radiochemical separations require a total of 16 man-hours for two samples. By running samples concurrently, one person can obtain final results on about 5 samples per week, involving 5 analyses for each sample, or a total of 25 analyses.

Counting

Counting time and sample capacities of the alpha and beta counters are shown in table 5.

Table 5. Sample capacity of alpha and beta counters

Counter	Number	Unit sample capacity	Counting duration (min)	Sample capacity/week at NERHL
Beta:				
Low-background automatic changer	2	50	30	[a] 220
Alpha:				
Internal proportional counter	4	1	100	[b] 78

[a] 20 hr/week are allowed for background counting, calibration, and sample changing.

[b] 30 hr/week are allowed for background counting, calibration, and sample changing.

Calculations

Gamma spectrum punchtapes are analyzed by computer techniques whereas chemical and radiochemical results are hand calculated by statistical methods. Each calculation is doublechecked for errors. Time requirements for calculation are listed in table 6.

Table 6. Calculation time requirements

Type of analysis and nuclide	Man-hours required		Total man-hours/ calculation
	Original calculation	Calculation check	
Gamma:			
Cesium 137, Cerium 141–144, Zinc 65, Zirconium 95–Niobium 95, Barium 140–Lanthanum 140, Ruthenium 103–106, Iodine 131, Potassium 40	0.133 (computer)	—	0. 133
Beta:			
Strontium 90	0. 117	0. 100	0. 217
Strontium 89	0. 117	0. 100	0. 217
Alpha:			
Total Radium	0. 117	0. 100	0. 217

SUMMARY

The estimation and assessment of the human intake of radioactive materials require the analysis of a wide variety of food items, using techniques capable of very low limits of sensitivity. The securing of meaningful data for evaluation of nationwide trends necessitates the processing of large numbers of samples. Facilities for receiving, storing, and processing of such samples have already been described. Analytical procedures must be as exacting as possible; thus various steps in the processing are time-consuming. For accuracy of results, analytical equipment must be carefully calibrated. In addition, a program of quality control on the accuracy, reliability, and reproducibility of analytical procedures performed on reference samples placed in the assembly line is particularly important to an effective large-scale analysis system.

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SECTION II

ROUTINE ANALYSIS OF RADIONUCLIDES IN WATER

Paul J. Magno and Lloyd R. Setter

INTRODUCTION

As part of its responsibilities for protecting the public against the harmful effects of ionizing radiation, the Division of Radiological Health, Public Health Service, in cooperation with Federal agencies and State and local health departments, conducts surveillance programs to determine the presence and extent of radioactive material in the environment. An important phase of these programs is the determination of gross activity and specific radionuclide activities in fresh and saline waters, for purposes of measuring the human intake through ingestion and determining radioactive material released to ground and surface waters. Specific examples include the surveillance of port environs in which nuclear submarines and ships operate, and the evaluation of aquatic systems receiving radioactive wastes discharged from large nuclear installations.

In keeping with the Division's interests in these areas, personnel of the Division have developed analytical methods for the measurement of levels of radioactive materials in water. Because of the many parameters associated with the environmental aspects of water and the low levels of radioactivity generally associated with it, numerous samples are analyzed at the various regional laboratories of the Division where the necessary processing techniques have been adapted to mass production. The manner in which water samples are examined on a mass analysis basis at the Northeastern Radiological Health Laboratory is described in this report.

ORGANIZATIONAL ASPECTS

In establishing an analysis system for the determination of radionuclides in environmental water, there are two criteria of importance: A large number of samples must be processed routinely and quickly, and the results obtained must be accurate. The system described in this report represents what is considered a satisfactory balance of these two requirements for surveillance purposes.

In order to facilitate the handling of large numbers of samples, the analytical-service group at the laboratory is divided into five sections:

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1. Sample Preparation.
2. Counting Laboratory.
3. Chemistry Laboratory.
4. Statistics.
5. Quality Control.

Figure 1 shows the flow of samples through the laboratory.

All samples arriving at the laboratory are received at the sample-preparation room. Here they are logged in and assigned identification numbers. In order to facilitate the automatic processing of the sample data, a six-digit number is assigned. The first two digits are code numbers categorizing the sample, the third digit indicates the number of times the sample is to be analyzed (duplicate, triplicate, and so forth), and remaining digits identify the sample. An information card containing necessary information, such as collection date, volume of sample, and unit results desired, accompanies the sample in its course through the laboratory.

The sample is first submitted to the counting laboratory for gamma analysis. These data are recorded on punched tape which in turn is fed to a computer for calculation of the concentration of the gamma-emitting radionuclides in the sample.

The sample is then submitted to the chemistry laboratory for gross alpha and beta analyses and any other specific determinations required or requested. Data from these analyses are submitted to a statistical group for calculation and reporting. The remaining portion of the sample is returned to the sample-preparation room and stored for 6 months in the event a rerun of the sample is requested.

To assure the reliability of analytical results, an intralaboratory quality control program is maintained. Twenty percent of the routine samples, selected randomly, are resubmitted for analysis. The identity of these control samples is unknown to the analyst. This number of blind duplicates permits evaluation of the reproducibility of analytical results. The agreement between samples is measured by statistical techniques that compare the observed differences between duplicates with the variability expected in the analyses. While reproducibility is one important measure of analytical validity, accuracy can be assured only when results are compared to those expected from a standard sample. Therefore, in addition to the blind duplicates, samples of known activity are periodically submitted for analysis and compared statistically to the expected values (*1*).

Since the counting equipment serves such an important function in the analyses, a quality control program involving daily checks on the background and counting efficiencies is also maintained.

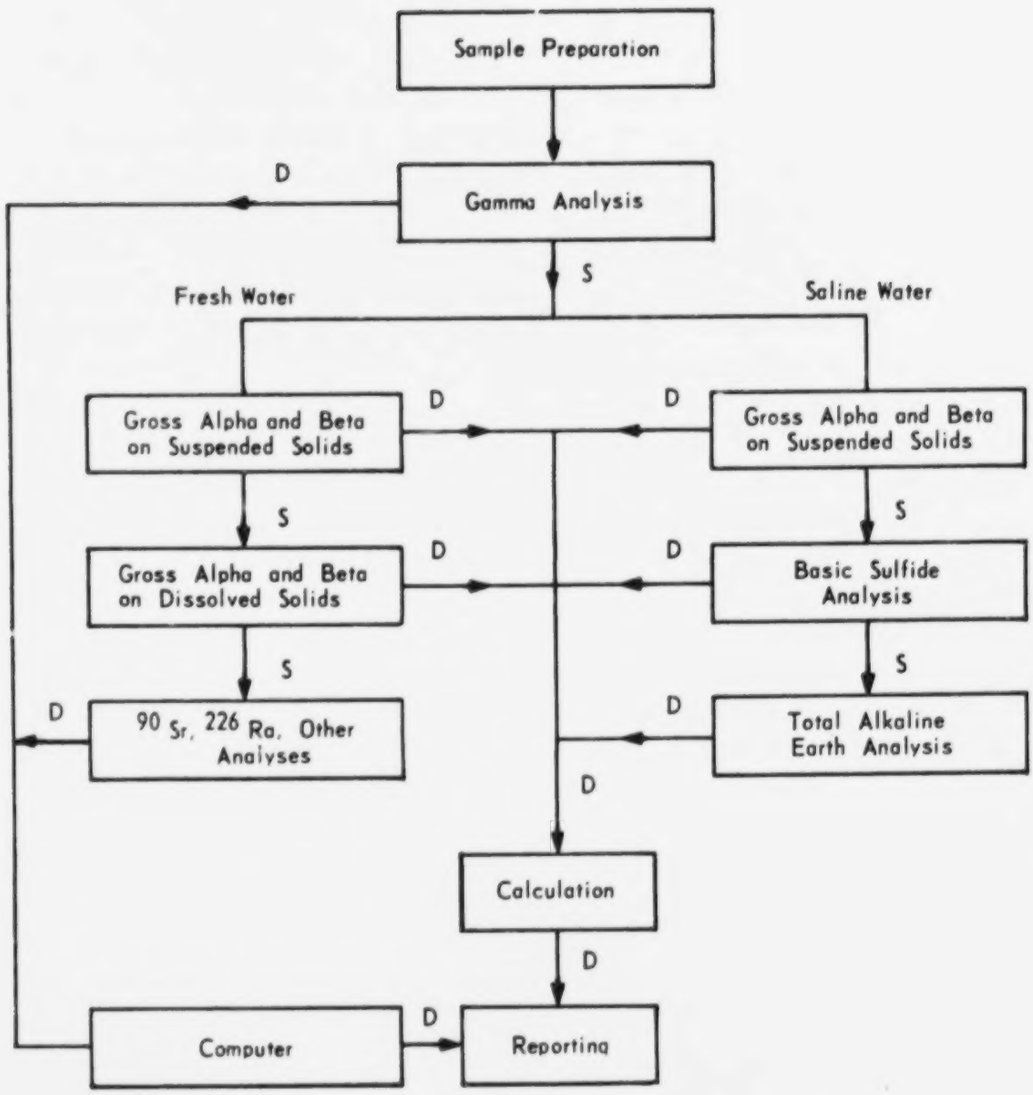

Figure 1. Flowchart for water sample processing

TECHNICAL ASPECTS

GAMMA ANALYSIS

Gamma analysis is carried out using a 4- by 4-inch NaI(Tl) crystal in a shielded chamber with a multichannel pulse height analyzer. A 3.5-liter sample in a Marinelli beaker is counted for 50 minutes. The gamma-emitting radionuclides [1] usually determined in water are cesium 137, cerium 144, manganese 54, zinc 65, zirconium-niobium 95, barium-lanthanum 140, ruthenium 103–106, iodine 131, and potassium 40. The counting equipment is calibrated for each radionuclide to be determined or known to be present. From these calibrations a series of matrix equations is prepared in a manner similar to those equations described by Hagee *et al.* (*2*) for milk. These equations allow the calculation of the concentration of each radionuclide in the presence of a mixture. Table 1 lists the detectable limits for the radionuclides given above.

Table 1. Detection limits for gamma-emitting radionuclides in water [a]

Radionuclide	pCi/liter
Barium-Lanthanum 140	8
Cerium 144	58
Cesium 137	12
Iodine 131	9
Potassium 40	80
Manganese 54	8
Ruthenium 103–106	50
Zinc 65	15
Zirconium-Niobium 95	8

[a] Using a 3.5-liter sample in a Marinelli beaker.

RADIOCHEMICAL ANALYSIS

After gamma analysis, the samples are submitted to the chemistry laboratory where they are separated into two categories—fresh and saline water. For the purposes of this laboratory, samples containing less than 1,000 ppm of dissolved solids are considered to be fresh water.

[1] Other radionuclides, if present, may be measured by proper calibration of the equipment.

Fresh water samples are analyzed for gross alpha and beta activity in both the suspended and dissolved fractions (*3*). These gross counting data are evaluated with respect to the origin of the sample and the gamma analysis data. In general, specific analysis for radium 226 is considered when the gross alpha count on a drinking-water sample exceeds 3 pCi/liter and for strontium 90 when the gross beta count on a drinking-water sample exceeds 10 pCi/liter. These limits are in accordance with the recommendations given in the Public Health Service Drinking Water Standards (*4*).

The suspended solids fraction of the saline-water samples is also analyzed for gross alpha and beta activity; however, because of the self-absorption effects, the measurement of gross alpha and beta of dissolved solids in saline water is impractical and is not done. However, basic sulfide and total alkaline earth separations allow for group measurements of most of the important radionuclides in saline water. A basic sulfide precipitate contains activity from many of the important fission products and reactor corrosion products, including radionuclides of the rare earths, chromium, zinc, ruthenium, zirconium, cerium, and cobalt. Uranium, thorium, and plutonium would also be included in this precipitate. The total alkaline earth precipitate contains the radionuclides of strontium, barium, and radium.

A brief description of the procedures used for these analyses is presented below:

Fresh Water

Gross Alpha and Beta in Suspended Solids (5)

A sample of 1 liter or an aliquot containing less than 200 mg of suspended solids is filtered through a 2-inch-diameter membrane filter. The filter is dried, and the weight of the suspended solids is measured. The cellulose membrane filter is saturated with alcohol-acetone and ignited in a 2-inch-diameter planchet. The organic matter of the sample is destroyed at low temperatures by burning over a Meeker burner. The fixed solids are cooled, distributed uniformly over the planchet with the aid of distilled water and a spatula, dried on a hot plate, and weighed. The sample thickness is maintained below 10 mg/cm^2. The sample is counted for gross alpha activity for 50 minutes in an internal proportional counter, and for gross beta activity for 30 minutes in a low-background beta counter.

Gross Alpha and Beta in Dissolved Solids—Fresh Water (5)

An aliquot of the filtrate from the suspended solids analysis containing up to 200 mg of dissolved solids (determined by measuring the conductivity of the solution) is evaporated to a small volume, trans-

ferred with washings to a 2-inch-diameter planchet, and dried in an oven at 105° C. The weight of dissolved solids is determined and the planchet counted for gross alpha activity for 50 minutes in an internal proportional counter and for gross beta activity for 30 minutes in a low-background beta counter.

Strontium 90 (6)

Strontium carrier is added to 1 liter of a filtered freshwater sample and the alkaline earths are precipitated as the carbonates. The strontium is separated by precipitation as the nitrate and further purified by barium chromate precipitation and hydroxide scavenging. The strontium is then precipitated as the carbonate, dissolved in dilute HNO_3, and stored after adding yttrium carrier. Yttrium carrier, including ingrown yttrium 90, is then extracted with 2-thenoyltrifluoroacetone (TTA) from the aqueous phase at pH 5 and back-extracted with dilute nitric acid. The yttrium 90 is then precipitated as the oxalate, weighed to determine yield, and counted for 30 minutes for yttrium 90 beta activity in a low-background beta counter. Figure 2 shows a flow diagram of this procedure.

Radium 226 (7)

Barium carrier and barium 133 tracer are added to 1 liter of a filtered freshwater sample and the radium coprecipitated with barium sulfate. The precipitate is converted from the sulfate to the carbonate and dissolved in nitric acid. The barium yield is determined by gamma counting the barium 133. The solution is deemanated and aged for 2 weeks to allow for ingrowth of radon 222. The solution is again deemanated, the radon collected in a scintillation cell and the alpha activity measured in a radon counter for 50 minutes. Figure 3 shows a flow diagram of this procedure.

Saline Water

Gross Alpha and Beta Suspended Solids

This procedure is the same as for Fresh Water.

Basic Sulfide Analysis—Saline Water (8)

Iron, cobalt, zinc, nickel, lanthanum, and strontium carriers are added to the filtrate from the suspended solids analysis. The solution is acidified, boiled, and made basic with ammonium hydroxide. Hydrogen sulfide is passed through the solution and the precipitate is filtered, washed and ignited. The ash is transferred to a 2-inch-diameter planchet and counted for gross alpha activity for 50 minutes in an internal proportional counter, and for gross beta activity for 30 minutes in a low-background beta counter.

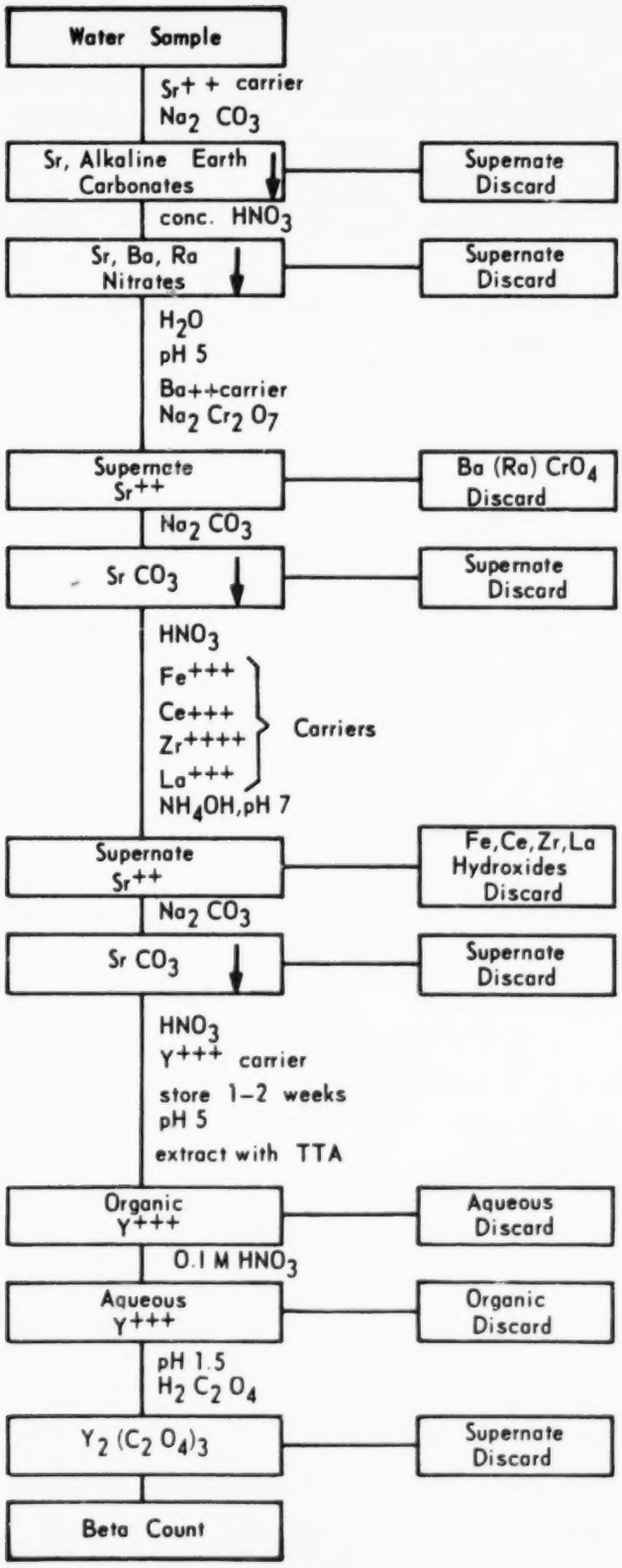

Figure 2. Flow diagram of procedure for strontium 90 analyses in water

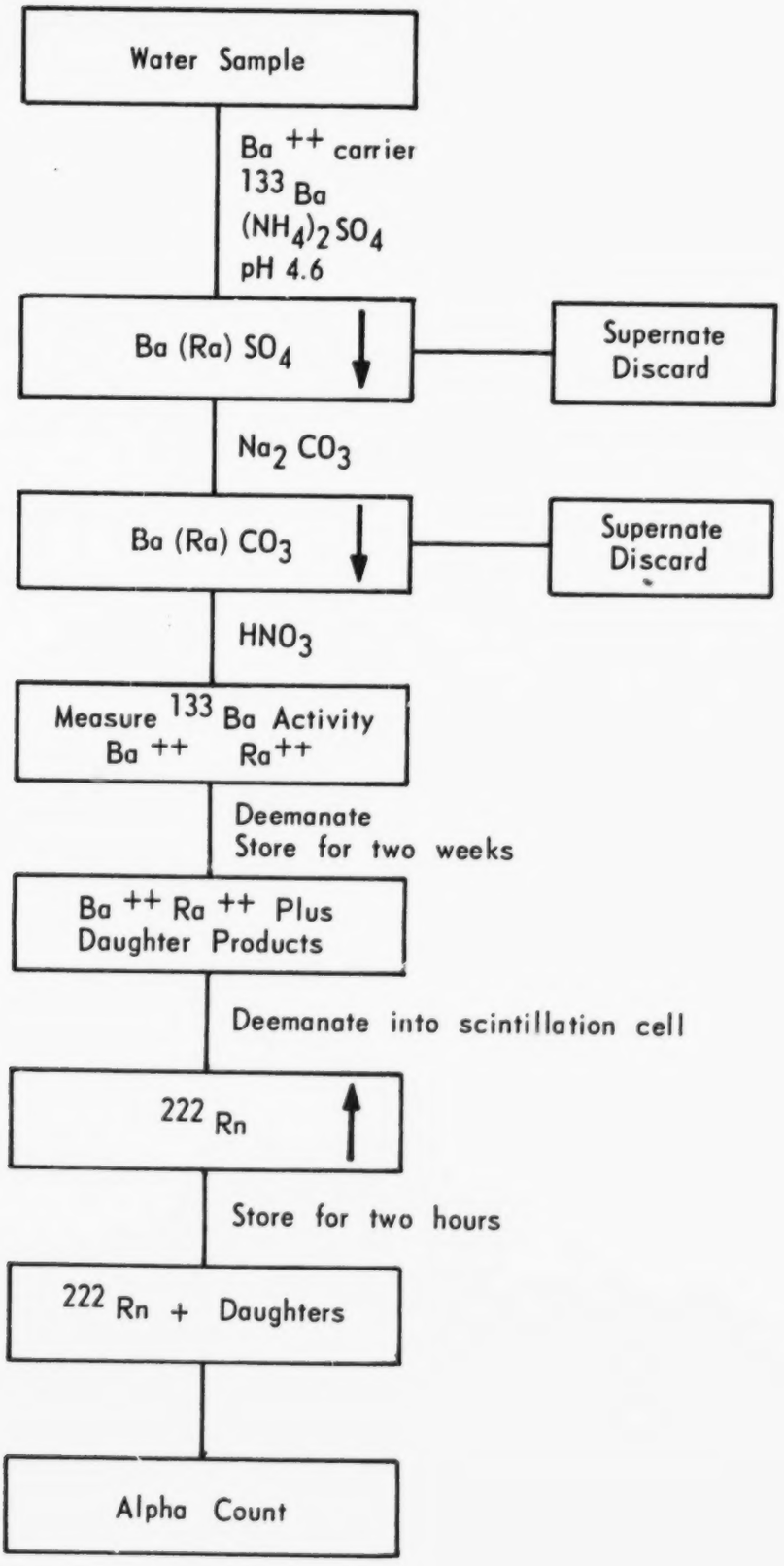

Figure 3. Flow diagram of emanation procedure for the analyses of radium 226 in water

Total Alkaline Earth Analysis—Saline Water (8)

The alkaline earths are precipitated as carbonates from the filtrate from the basic sulfide analysis. The carbonates are dissolved in dilute nitric acid, and strontium, barium, and radium are precipitated as nitrates free of calcium. The precipitate is dissolved in water and then precipitated as carbonates that are mounted in a 2-inch-diameter planchet and counted for gross alpha activity for 50 minutes in an internal proportional counter, and for gross beta activity for 30 minutes in a low-background beta counter.

Calculations

$$\text{Gross } \alpha \text{ and } \beta \text{ (pCi/liter)} = \frac{\text{cpm}}{2.22 \times \text{Efficiency} \times \text{Sample Volume (liters)}}.$$

Calibration curves relating the efficiency factors to the weight of solids present are used in calculating the gross alpha [1] and beta activities.[2]

$$\text{Strontium 90 (pCi/liter)} = \frac{A}{2.22\ B\ C\ D\ E\ F\ G}.$$

A = Net counts per minute yttrium 90.
B = Sample aliquot in liters.
C = Yttrium 90 efficiency factor.
D = Yttrium chemical yield.
E = Yttrium 90 decay factor.
F = Yttrium 90 ingrowth factor.
G = Strontium chemical yield.

$$\text{Radium 226 (pCi/liter)} = \frac{A}{2.22\ B\ C\ D\ E\ F}.$$

A = Net counts per minute.
B = Sample volume in liters.
C = Efficiency of deemanation and counting.
D = Barium 133 yield.
E = Radon 222 ingrowth factor.
F = Radon 222 daughter ingrowth factor.

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[1] Natural uranium is used as an alpha calibrating source.
[2] Thallium 204 and cesium 137 are the preferred gross beta calibrating sources (*5, 9, 10*), while strontium-yttrium 90 is reasonably satisfactory provided the combined sample and counter window thickness is restricted preferably below 5 mg/cm^2.

OPERATIONAL ASPECTS

MANPOWER REQUIREMENTS

Table 2 lists the manpower requirements, in man-hours per sample, for the various phases of the analyses, and also shows the elapsed time required to complete each phase, assuming continuous processing from the time the sample is received at the laboratory.

As can be seen, gamma spectrometry provides a simple and rapid analytical technique requiring a minimum of manpower. However, it must be noted that this technique is applicable to relatively simple mixtures of gamma-emitting nuclides (usually 10 or less) and that the amount of activity present must exceed those values listed in table 1 to be detected. On the other hand, gross alpha and beta measurements are more sensitive but do not discriminate among radionuclides present and are somewhat more time-consuming than gamma spectrometry. Radiochemical analyses provide a means of separating radionuclides, such as strontium 90 and radium 226, from each other, and from other less important radionuclides but at the expense of manpower and elapsed time requirements.

Table 2. Manpower requirements for analyses of radionuclides in water

Operation	Man-hours/ sample	Elapsed time (hours)
Sample preparation	0.1	0.1
Gamma analysis	[a] 0.3	[b] 1.0
Gross alpha and beta analysis/suspended solids	0.4	[c] 4–24
Gross alpha and beta analysis/dissolved solids	0.4	6
Radiochemical separations:		
Basic sulfide	1.0	8
Total alkaline earths	1.5	6
Strontium 90	6.0	10 days (7 days ingrowth)
Radium 226	4.0	16 days (14 days ingrowth)

[a] The time requirement for a gamma analysis is based on the use of a computer to make the calculations. Manual calculation of the concentrations of radionuclides in samples would increase the man-hour requirement to 1.0 man-hour/sample.

[b] This value is based on immediate availability of computer facilities. Since this is not usually possible the elapsed time for obtaining final results of this analysis could be as much as 1 week.

[c] Time required for filtering a sample is dependent upon the amount of suspended solids present and the filtering characteristics.

EQUIPMENT REQUIREMENTS

Pertinent data on the counting equipment necessary to conduct the various analyses are presented in table 3. The data include the capacity for each counter based on a 40-hour work week. Approximately 20 percent of the equipment capacity is used for quality control purposes in conducting routine checks on counter background and efficiency.

In the analysis of radium 226, a 50-minute background count is taken on the scintillation cell prior to the radon measurement. This increases the total counting time requirement for each analysis to 100 minutes and reduces the counter capacity by a factor of 2.

Other special equipment necessary to carry out the analyses includes a "radon bubbler" used for storage of the sample during radon ingrowth and for deemanation of the radon from the samples and scintillation cells used for counting the radon.

The use of automatic data processing systems greatly simplifies the computation of results from the raw data obtained by gamma spectrometry. The requirements for use of such a system are:

1. A means of producing from the gamma spectrometer unit an output which is suitable for computer input, that is, punched cards, punched tape, or magnetic tape. The laboratory uses punched paper tape.
2. Availability of a computer. The laboratory uses the IBM 1620 computer located at the Division of Radiological Health's laboratory in Rockville. Tapes are submitted and the results returned by mail. (This usually requires 1 week.)
3. Computer programs to perform the computations. The laboratory uses the programing capabilities of the Computer Group at the Rockville Radiological Health Laboratory, Rockville, Md.

Table 3. Counting equipment for analysis of radionuclides in water

Equipment	Counting time (min)	Capacity (samples/ week)[a]
400-channel analyzer split into two 200-channel analyzers, used with 4-by 4-inch NaI (Tl) crystal	50	80
Low-background beta counter with automatic sample changer, 2-inch detector	30	[b] 240
Internal alpha proportional counter, chamber for 2-inch-planchet, background 1 cph	50	40
Radon counter for radium 226 analysis	50	20

[a] Assuming a 40-hour week.
[b] Based on a 5-day week, 24 hour a day operation.

SUMMARY

Radioactive contamination of drinking water and of ground and surface water systems has been under surveillance by the Public Health Service for a number of years (*7*). To assure that the intake of radioactive materials through the ingestion of drinking water, and the levels of radioactivity in the aquatic environment can be reliably and accurately assessed, the Division of Radiological Health has established a system of analytical procedures for each of the more important determinations. Design of these procedures has been based on the need for processing numerous samples generally containing low levels of radionuclides.

In addition to gross alpha and beta measurements, the samples are examined by gamma spectrometry for specific nuclides, such as cesium 137, zinc 65, zirconium-niobium 95, and ruthenium 103, and by radiochemical separation for nuclides, such as radium 226 and strontium 90. Separation techniques are utilized in the preparation of saline waters for gross counting since these samples contain such large concentrations of dissolved solids as to make the more routine procedures unacceptable. To assure precision in the resulting data, quality control procedures have been integrated into the overall analytical process and include the analysis of blind duplicates and standard samples.

Radiochemical procedures for the determination of specific radionuclides are most demanding in terms of time and manpower. Automation of the gamma-counting equipment and the use of computers in the evaluation of associated spectral data have contributed significantly to the development of an instrumental approach requiring a minimum of manpower. By the proper use of a combination of both instrumental and radiochemical procedures, coupled with careful laboratory management, the Northeastern Radiological Health Laboratory has developed an effective system for the processing of large numbers of water samples.

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